REST AVAILABLE COPY

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date 1 November 2001 (01.11.2001)

PCT

(10) International Publication Number WO 01/82399 A1

(51) International Patent Classification7:

.

(21) International Application Number: PCT/US00/23043

(22) International Filing Date: 22 August 2000 (22.08.2000)

(25) Filing Language:

English

H01M 8/02

(26) Publication Language:

MN 55133-3427 (US).

English

(30) Priority Data: 09/557,712

25 April 2000 (25.04.2000) Us

(71) Applicant: 3M INNOVATIVE PROPERTIES COM-PANY [US/US]; 3M Center, P.O. Box 33427, Saint Paul,

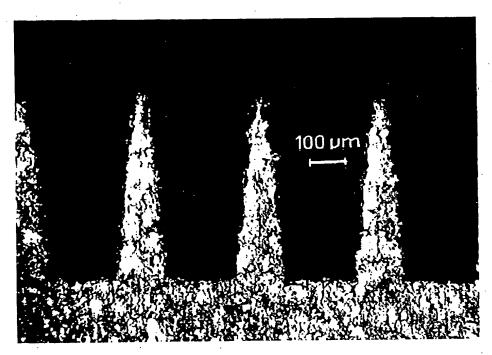
(72) Inventors: LEWINSKI, Krzystof, A., P.O. Box 33427,
 Saint Paul, MN 55133-3427 (US). FLEMING, Patrick,
 R.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US).

FEHR, Robert, T.; P.O. Box 33427, Saint Paul, MN 55133-3427 (US).

- (74) Agents: DAHL, Philip, Y. et al.; Office of Intellectual Property Counsel, P.O. Box 33427, Saint Paul, MN 55133-3427 (US).
- (81) Designated States (national): AE, AG, AL, AM, AT, AT (utility model), AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CR, CU, CZ, CZ (utility model), DE, DE (utility model), DK, DK (utility model), DM, DZ, EE, EE (utility model), ES, FI, FI (utility model), GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KR (utility model), KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SK (utility model), SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW.
- (84) Designated States (regional): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE,

[Continued on next page]

(54) Title: CONDUCTANCE ENHANCEMENT MICROSTRUCTURES FOR BIPOLAR PLATES



(57) Abstract: A bipolar plate for use in an electrochemical cell such as a hydrogen fuel cell is provided, comprising electrical contact surfaces and fluid conducting channels, wherein at least one of the electrical contact surfaces comprises a plurality of contact-enhancing microstructures.

01/87399 A1

BEST AVAILABLE COPY

WO 01/82399 A1



IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

Published:

with international search report

CONDUCTANCE ENHANCEMENT MICROSTRUCTURES FOR BIPOLAR PLATES

5

15

20

25

30

Field of the Invention

This invention relates to bipolar plates useful in electrochemical cells such as fuel cells. The bipolar plates of the present invention comprise fluid conducting channels separated by electrical contact surfaces, where the electrical contact surfaces bear microstructured features which enhance electrical conductance.

Summary of the Invention

The present invention provides a bipolar plate (BPP) for use in an electrochemical cell, the BPP comprising electrical contact surfaces and fluid conducting channels or fluid-conducting zones, wherein at least one of the electrical contact surfaces comprises a plurality of contact-enhancing microstructures.

In another aspect, the invention provides a two-layer structure comprising a bipolar plate bearing contact-enhancing microstructures and a diffuser/current collector (DCC).

It is an advantage of the present invention to provide a bipolar plate for improved fuel cell performance resulting from improved electrical conductivity. It is a further advantage of the present invention to provide a BPP-DCC combination having improved electrical conductivity.

Brief Description of the Figures

Figure 1 is a graph of resistance vs. pressure across one bipolar plate-DCC interface according to the present invention and one comparative interface.

Figure 2 is a scanning electron micrograph of an electrical contact surface bearing contact-enhancing microstructures according to the present invention. The micrograph includes a 1mm scale bar.

Figure 3 is a light micrograph of an electrical contact surface bearing contact-enhancing microstructures according to the present invention. The micrograph includes a 100 µm scale bar.

Detailed Description of Preferred Embodiments

5

10

15

20

25

The present invention provides an improved bipolar plate (BPP) for an electrochemical cell such as a fuel cell. The bipolar plate of the present invention comprises electrical contact surfaces and fluid-conducting channels, wherein one or more of the electrical contact surfaces comprise a plurality of contact-enhancing microstructures, which are microstructures which decrease the electrical resistance between the BPP and an adjacent structure.

Electrochemical cells include fuel cells, sensors, electrolyzers, and electrochemical reactors. Fuel cells utilize a fuel such as hydrogen and an oxidizing agent such as oxygen to produce an electrical current. The two chemical reactants, i.e., the fuel and the oxidizing agent, separately react at two isolated electrodes containing catalyst. An ion exchange element is located between the electrodes to prevent direct reaction of the two reactants and to conduct ions. In the case of a typical hydrogen fuel cell, the ion exchange element is an ion conducting membrane (ICM). The ICM conducts protons (H[†]) from the hydrogen electrode to the oxygen electrode. Electrons follow a separate electrical path, thereby generating an electric current. The combination of an ICM and electrodes is commonly referred to as a "membrane electrode assembly," or MEA.

In conventional fuel cells, MEA's are arranged in a stack separated by rigid, electrically-conductive plates which may be known as bipolar plates (BPP). The bipolar plate has one or more fluid-conducting channels engraved, milled, or molded in the surface(s) facing the MEA(s). The fluid-conducting channels on one side of the plate direct fuel to the anode of one MEA while the channels on the other side direct oxidant to the cathode of the next MEA in the stack. The bipolar plates conduct the electrical current generated in each MEA throughout the stack. As used herein, "bipolar plate" should be understood to include the end plates of a stack, which perform the functions of the bipolar plate on one side only and

serve the top and bottom MEA's of the stack. A stack having a single MEA has only two end plates, which are both encompassed by the term "bipolar plate" as used herein.

An additional layer, the diffuser/current collector (DCC), may be situated between the bipolar plate and the active catalytic sites of the MEA. Like the bipolar plate, the DCC must conduct fluids and electricity to and from a catalyst surface of the MEA. Unlike the bipolar plate, typical DCC's are porous throughout and malleable rather than rigid. Also, the BPP conducts fluids principally in the XY plane whereas the DCC conducts fluids principally in the Z direction. The DCC preferably comprises carbon fiber paper such as ELATTM electrode backing material (E-tek, Inc., Natick, MA), preferably at a thickness of about 0.4 mm. Another preferred material is Toray Carbon Paper (Toray Industries, Inc., Tokyo, Japan).

5

10

15

20 -

25

30

The bipolar plate of the present invention is made of an electrically conductive material such as a metal or conductive carbon materials such as graphite. Graphite is preferred due to its resistance to corrosion. Alternately, materials may be used which are plated, sputtered or otherwise coated with conductive layers by wet methods, vacuum methods, or any suitable method.

The bipolar plate of the present invention comprises electrical contact surfaces, which are typically land areas between fluid conducting channels. The bipolar plate of the present invention comprises contact-enhancing microstructures borne on the electrical contact surfaces to enhance electrical contact between the bipolar plate and the DCC. It has been found that the use of such structures can reduce electrical resistance in a BPP-DCC combination. The microstructures may comprise any suitable shapes, including points, ridges, bumps, posts or the like. At least one electrical contact surface bears a plurality of such structures. The microstructures preferably have a height that is less than the thickness of the DCC with which they are used. The microstructures preferably have a height that is at least 1% of the thickness of the DCC. In another preferred embodiment the microstructures have a height that is at least 5% of the thickness of the DCC. In another preferred embodiment the microstructures have a height that is between 10% and 90% of the thickness of the DCC. As used herein, height of a microstructure refers to the difference in elevation, along an axis normal to the plane of the bipolar plate, of a local maximum at the peak of a microstructure and a local minimum between two directly adjacent microstructures. Preferably the

microfeatures have a height of less than 400 μm, more preferably less than 200 μm, more preferably less than 100 μm, and most preferably less than 50 μm. Preferably the microfeatures have a height of at least 10μm. Preferably the distance between microstructures or pitch of the microstructure pattern is less than 400 μm, more preferably less than 200 μm, more preferably less than 100 μm, and most preferably less than 50μm. As used herein, pitch means the smallest distance between analogous structures in a repeating pattern, e.g., peak-to-peak or edge-to-edge. Preferably, the microfeatures have an aspect ratio (ratio of height to pitch, as defined above) of at least 0.1, more preferably at least 0.5, and more preferably about 2. In another preferred embodiment, the microfeatures have an aspect ratio of greater than 2. In another preferred embodiment, the microfeatures have an aspect ratio of greater than 5. In another preferred embodiment, the microfeatures have an aspect ratio of greater than 10.

5

10

15

20

25

The bipolar plate of the present invention comprises fluid conducting channels for transport of reactants and products. The channels may conduct gasses, liquids or both; including fuels such as hydrogen, oxidizing agents such as air or oxygen, and products such as water. The channels may be of any suitable design, such as a series of parallel channels, one or more serpentine channels, a gridwork of intersecting channels, or the like. In a preferred embodiment, these microflow channels comprise a highly parallel pattern. These patterns may contain interconnections or branch points. Such patterns include parallel lines, hatching, and grid patterns of channels. The channels can be of any cross-sectional geometry that provides desired fluid transport, but preferably a geometery which is readily replicated. The channels terminate at one or both ends in an inlet, outlet or manifold to allow introduction or removal of fluid.

The channels may be conventional channels or, more preferably, the microstructured channels described in copending U.S. patent application serial number 09/430,568. In contrast to the microstructured flow field features of that application, the microstructures of the present invention are situated on the electrically conductive land features of the bipolar plate. The volumes lying between directly adjacent microstructured features according to the present invention do not conduct substantial amounts of any fluid. The volumes defined by adjacent microstructured features may lack substantial communication with any inlet, outlet

or manifolds; they may be substantially occluded by DCC material under conditions of use; and they are accompanied by channels, either conventional or microstructured, which do conduct substantial amounts of fluid.

The bipolar plates of the present invention may be made by any suitable method, including the methods described in U.S. Patent No. 5,728,446, to Johnston, et. al., and pending U.S. Patent Application S.N. 09/099,269, both of which are incorporated herein in full by reference.

5

10

15

20

25

30

The bipolar plates of the present invention preferably have fluid-conducting channels as described above, but may lack these channels in the case described following. Where the DCC adjacent to the bipolar plate has fluid-conducting channels cut, molded, or otherwise formed into it, the bipolar plate may not require fluid-conducting channels since this function is performed by the DCC. In that case, the surface area of the plate that is opposite a fluid-conducting channel of the DCC is a fluid conducting zone. Such a plate according to the present invention has contact-enhancing microstructures borne on the electrical contact surfaces and may or may not have microstructures on the plate surface constituting a fluid-conducting zone. The present invention contemplates a two-layer construction comprising a plate bearing contact-enhancing microstructures on electrical contact surfaces in contact with a DCC which comprises fluid-conducting channels.

This invention is useful in constructing electrochemical cells such as fuel cells.

Objects and advantages of this invention are further illustrated by the following examples, but the particular materials and amounts thereof recited in these examples, as well as other conditions and details, should not be construed to unduly limit this invention.

Examples

Three test plates were tested, each representing the land area of a bipolar plate with conductivity-enhancing microstructures according to the present invention. In examples of the invention, the microstructures were pressed against a DCC layer, which comprises carbon paper. For the comparative examples, each of the three test plates was reversed so that its flat back surface contacted the DCC layer. Thus, the comparative trials represent plates of exactly equal mass, composition and outer dimensions but lacking microstructure-DCC contact. All

plates were made of nickel metal. The test plates had dimensions of approximately 22.4 mm x 22.4 mm for an area of 5.0 cm². Figs. 2 and 3 are micrographs of a test plate made identically to those used in the present Examples.

5

10

15

20

25

Test plates 1, 2, 3 and the test plate appearing in Figs. 2 and 3 were cut from a large plate made using a nickel tool as described in U.S. Patent No. 5,728,446, Comparative Example 1, also described as pattern 2 of Table I. (U.S. Patent No. 5,728,446 is incorporated herein by reference.) The tool, which was a negative image of the plate, was first passivated by washing with a potassium dichromate solution. Nickel metal was electrodeposited from a solution of nickel sulfonate to form a positive image, which was the large plate. Nickel was deposited to a plate thickness of about 3.0 mm. The large plate was separated from the tool by hand and individual test plates were cut from it. The pattern of the test plates comprised parallel linear grooves 532 μm deep, on a 330 μm pitch, with wall angles for each groove of 5° from vertical and a peak top width of 29 μm.

Test plates 1, 2 and 3 were tested. Examples 1A, 2A and 3A are examples of the present invention and Examples 1C, 2C and 3C are comparative examples, performed with the flat side of the plate facing the DCC material. For each test plate, the face to be tested was placed against a layer of 0.404 mm thick DS (double sided) ELATTM electrode backing material (E-tek, Inc., Natick, MA), all ELAT sample being taken from the same lot. The two layer construction was then tested for electrical resistance through its thickness under varying degrees of compression, as described following. Each series of tests was performed with a new piece of ELAT.

Electrical resistance was measured as a function of compression pressure using an apparatus comprising a pneumatic press (Bellofram, State Route 2, Newell, West Virginia). The press was equipped with two parallel platens each having an area of 100cm^2 . The platen surface was polished brass and was electrically isolated from the rest of the press. The press includes a pressure meter with a digital output. The press includes a displacement meter (3 sensors, Sensotec, Columbus, OH) with a digital output serially interfaced to the computer. A regulated power supply (model HP 6269B, Hewlett-Packard, 3000 Hanover Street, Palo Alto, CA) was electrically connected to the platens so a fixed current could be applied between the

platens when a sample was in place. A voltmeter with a digital output (model Fluke 45 Dual Display Multimeter, Fluke Corporation, P.O. Box 9090, Everett, WA) was electrically connected to the platens so the voltage difference between the platens could be measured when a sample was in place. Data was collected from the pressure meter and voltmeter using a serial hardware interface and Lab View data acquisition software (National Instruments Corporation, 11500 N Mopac Expwy, Austin, TX).

5

10

15

20

25

For the resistance measurements, each of the plates were sandwiched between steel shims, with a DCC layer against one face, and pressed between the platens of the press. The voltage drop across each plate/DCC sample was measured as a function of applied pressure under a constant current of 2 A/cm². Current loads as high as 20 A/cm² were also tested to verify the data. It was found that at maximum pressure the value of measured resistance was independent of current density.

Fig. 1 is a graph of resistance vs. pressure for Examples 3A (Trace A) and 3C (Trace C). The data indicate that the microstructure-DCC contact arrangement demonstrated consistently lower resistance in comparison with the flat surface-DCC contact arrangement. Furthermore, upon release of pressure to less than 1.4 MPa (200 PSI) the microstructure-DCC contact sample retained its low resistance, whereas the flat surface-DCC contact sample recovered some of its resistance.

Table I presents resistance measurements for Examples 1A, 2A, 3A and an average of the three and for Examples 1C, 2C, 3C and an average of the three. The given measurements are for total resistance ($m\Omega^*cm^2$). Each resistance is the average of at least 30 data points gathered at the peak pressure, about 17 MPa (2500 PSI). Also presented is the standard deviation of each set of the last 30 data points. Table I also reports the difference in resistance between the examples of the invention and corresponding comparative examples, with standard deviation again given.

5

10

Table I

Sample	Resistance (mΩ*cm²)	S.D. $(m\Omega^*cm^2)$	Sample	Resistance $(m\Omega*cm^2)$	S.D. $(m\Omega^*cm^2)$	Difference $(m\Omega^*cm^2)$	S.D. $(m\Omega^*cm^2)$
1A	20.14	0.032	1C	22.29	0.038	2.15	0.050
2A	19.70	0.028	2C	22.33	0.037	2.63	0.047
3A	19.60	0.029	3C	24.06	0.043	4.47	0.051
Ave.	19.81	0.030	Ave.	22.89	0.039	3.08	0.049

The data in Table I again demonstrate that the microstructure-DCC contact arrangement had consistently lower electrical resistance in comparison with the flat surface-DCC contact arrangement.

Various modifications and alterations of this invention will become apparent to those skilled in the art without departing from the scope and principles of this invention, and it should be understood that this invention is not to be unduly limited to the illustrative embodiments set forth hereinabove.

We claim:

٠5 -

20

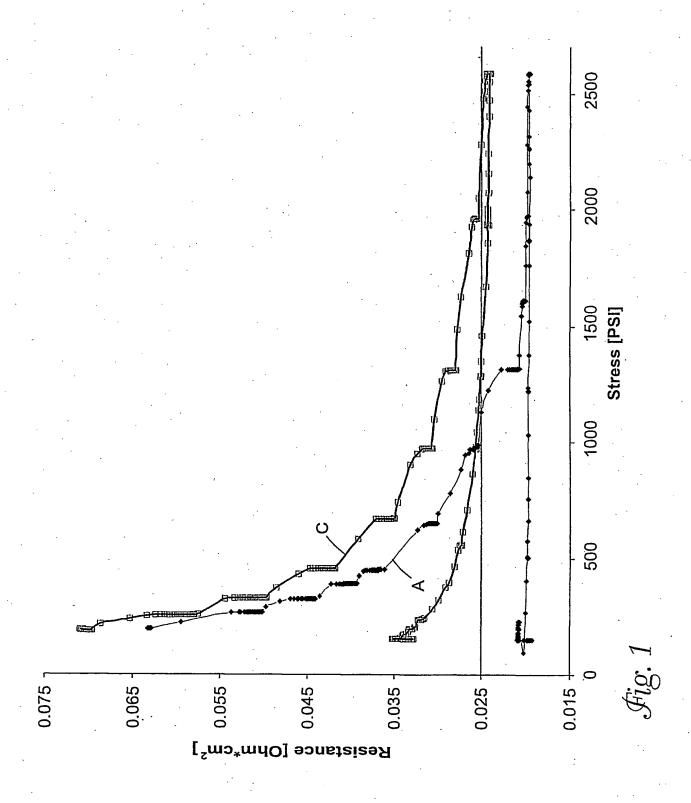
30

1. A bipolar plate comprising electrical contact surfaces and one of: fluid conducting channels and fluid-conducting zones; wherein at least one of said electrical contact surfaces comprises a plurality of contact-enhancing microstructures.

- 2. The bipolar plate according to claim 1 comprising fluid conducting channels.
- The bipolar plate according to claim 1 or 2 wherein said contact-enhancing
 microstructures have a pitch of less than 100 μm.
 - 4. The bipolar plate according to claim 1, 2 or 3 wherein said contact-enhancing microstructures have a height of less than 100 μm.
- 15 5. The bipolar plate according to claim 1, 2, 3 or 4 wherein said contact-enhancing microstructures have an aspect ratio of greater than 0.5.
 - 6. The bipolar plate according to claim 1, 2, 3 or 4 wherein said contact-enhancing microstructures have an aspect ratio of about 2.
 - 7. A two-layer construction comprising the bipolar plate according to any of claims 1 6 and a diffuser/current collector (DCC), wherein the diffuser/current collector (DCC) is in electrical contact with at least one of said electrical contact surfaces.
- 25 8. A two-layer construction according to claim 7 wherein said contact-enhancing microstructures have a height that is between 10% and 90% of the thickness of the DCC.
 - 9. A two-layer construction according to claim 7 or 8 wherein the volumes lying between directly adjacent contact-enhancing microstructures fulfill a condition selected from a) b) and c):

PCT/US00/23043

- a) they do not conduct substantial amounts of any fluid;
- b) they lack substantial communication with any inlet, outlet or manifold; and
- c) they are substantially occluded by DCC material under conditions of use.
- 5 10. A two-layer construction according to claim 7, 8, or 9 wherein the diffuser/current collector (DCC) is coated with an electrically non-conductive hydrophobic polymer.
- A two-layer construction comprising the bipolar plate according to any of claims 1 6 wherein said bipolar plate comprises fluid-conducting zones and a diffuser/current collector (DCC), wherein said diffuser/current collector (DCC) comprises fluid-conducting channels, and wherein said diffuser/current collector (DCC) is in electrical contact with at least one of said electrical contact surfaces.



2/3

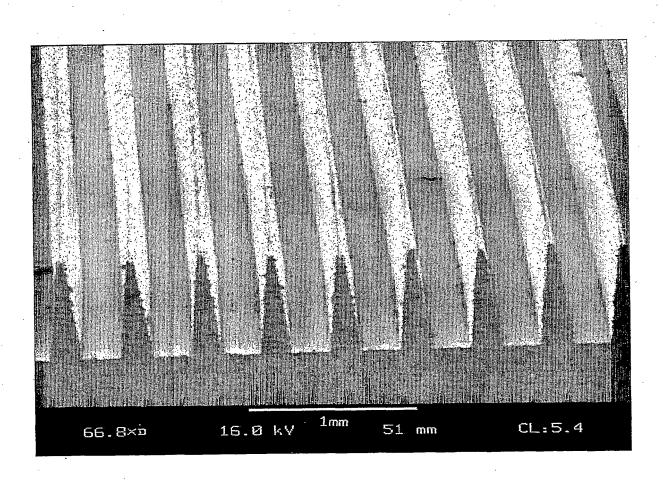


Fig. 2

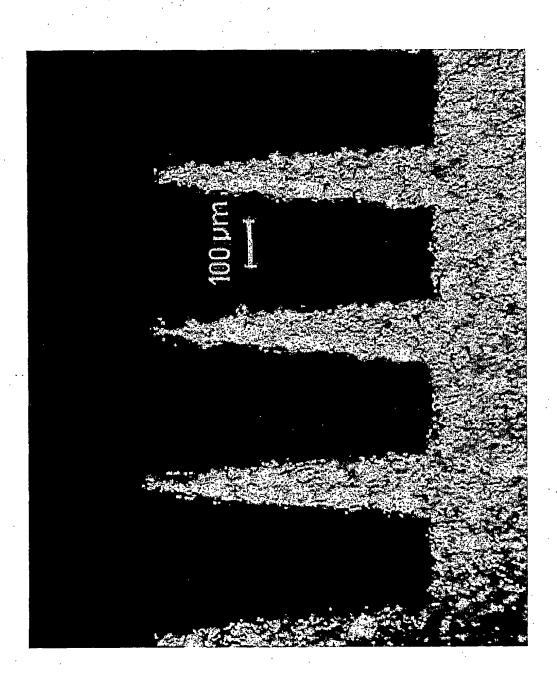


Fig. 3

BEST AVAILABLE COPY

INTERNATIONAL SEARCH REPORT

Interna Application No PCT/US 00/23043

	·		
a. classif IPC 7	ICATION OF SUBJECT MATTER H01M8/02	- -	
110 /	1102.10, 02		
A coording to	International Patent Classification (IPC) or to both national classifica	ation and IPC	
B. FIELDS S			
Minimum doc	cumentation searched (classification system followed by classification	on symbols)	
IPC 7	HO1M		
5 /	ion searched other than minimum documentation to the extent that s	uch documents are included in the fields sea	rched
Documentári	on searched other man minimum documentation of the		
Electronic da	ata base consulted during the international search (name of data ba	se and, where practical, search terms used)	
	ternal, PAJ, WPI Data		
o pooline	ENTS CONSIDERED TO BE RELEVANT		
Category °	Citation of document, with indication, where appropriate, of the re	levant passages	Relevant to claim No.
Jalegury	Ontarior of doddmining man and appropriately		<u> </u>
P,X	WO 00 26981 A (3M INNOVATIVE PRO CO) 11 May 2000 (2000-05-11) page 5, line 17 - line 24; claim	•	1-4
Α	US 3 814 631 A (WARSZAWSKI B ET 4 June 1974 (1974-06-04)		1-11
Α	US 5 683 828 A (MUEGGENBURG H HA AL) 4 November 1997 (1997-11-04) claims 1-25	RRY ET	1-11
А	WO 94 11912 A (BALLARD POWER SYS 26 May 1994 (1994-05-26) claims 1-42	TEMS)	1-11
ļ	Name parts state state		•
	·	·	
Fur	ther documents are listed in the continuation of box C.	χ Patent family members are listed	in annex.
° Special c	eategories of cited documents:	"T" later document published after the inte	rnational filing date
'A' docum	nent defining the general state of the art which is not	or priority date and not in conflict with cited to understand the principle or the	the application out
consi	idered to be of particular relevance cocument but published on or after the international	invention -	laimed invention
filing	date	cannot be considered novel or cannot involve an inventive step when the do	pe considered to
l which	nent which may throw doubts on priority claim(s) or h is cited to establish the publication date of another	"Y" document of particular relevance; the cannot be considered to involve an in	laimed invention
"O" docum	on or other special reason (as specified) nent referring to an oral disclosure, use, exhibition or	document is combined with one or ments, such combination being obvio	ore other such docu-
other	r means means published prior to the international filling date but than the priority date claimed	in the art. *& document member of the same patent	
1	e actual completion of the international search	Date of mailing of the international se	
	22 December 2000	03/01/2001	
<u> </u>	malling address of the ISA	Authorized officer	
value and	European Patent Office, P.B. 5818 Patentlaan 2		•
	_NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Battistig, M	

INTERNATIONAL SEARCH REPORT

ination on patent family members

Interna Application No
PCT/US 00/23043

Patent document cited in search report			Publication date	Patent family member(s)		Publication date	
WO	0026981	Α .	11-05-2000	AU	1600800 A	22-05-2000	
US	3814631	Α	04-06-1974	FR	2125159 A	29-09-1972	
	•		÷	DE	2205294 A	31-08-1972	
	•		•	GB	1373832 A	13-11-1974	
•				JP	56044545 B	20-10-1981	
				US	3901731 A	26-08-1975	
us	5683828	Α	04-11-1997	AU	4193996 A	06-05-1996	
	••••			CA	2202380 A	25-04-1996	
				EP	0783770 A	16-07-1997	
•				JP .	10507573 T	21-07-1998	
•				WO.	9612316 A	25-04-1996	
				US	5863671 A	26-01-1999	
			•	US	5858567 A	12-01-1999	
				US	6051331 A	18-04-2000	
	·					10 04 2000	
MC	9411912	Α	26-05-1994	AU	5415994 A	08-06-1994	
WO	, , , , , , , , , , , , , , , , , , , ,	,,		ÜS	5300370 A	05-04-1994	

THIS PAGE BLANK (USPTO)